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TITLE DEVELOPMENT OF NEUTRON MULTIPLICITY COUNTERS FOR SAFEGUARDS ASSAY

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DEVELOPMENT OF NEUTRON MULTIPLICITY COUNTERS FOR SAFEGUARDS ASSAY

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ABSTRACT

This paper reports on the development of a new generation of neutron multiplicity counters for assaying impure plutonium. The new counters will be able to obtain three measured parameters from the neutron multiplicity distribution and will be able to determine sample mass, multiplication, and (α,n) reaction rate, making it possible to obtain a more matrix-independent assay of moist or impure materials. This paper describes the existing prototype multiplicity counters and evaluates their performance using assay variance as a figure of merit. The best performance to date is obtained with a high-efficiency, low die-away-time thermal neutron counter with shift-register electronics.

I. INTRODUCTION

Neutron coincidence counters are used to verify or account for plutonium and uranium samples present in the nuclear fuel cycle. The counters are used for assay of scrap and waste, shipper/receiver confirmatory measurements, and inventory verification. A fundamental limitation of the current generation of neutron coincidence counters is their ability to measure only two parameters: the total neutron count rate T and the real coincidence count rate R. For a typical sample, there are at least three unknowns: mass m, neutron leakage multiplication M, and the fraction α of neutrons from (α,n) reactions relative to those from spontaneous fission. (If the sample contains neutron-moderating materials, then the neutron detection efficiency ε can be a fourth unknown.) Thus it is often not possible to obtain accurate assays of impure samples for which M and α are both unknown, and it is customary to use slower but more accurate techniques, such as calorimetry, for accountability of bulk plutonium samples.

The new neutron multiplicity counters under development are distinguished from existing neutron coincidence counters by their ability to measure a third parameter. The multiplicity counters measure the multiplicity distribution of neutrons emitted by the sample (the number of 0's, 1's, 2's, etc., registered after an initial trigger event). From the measured multiplicity distribution it is possible to extract three measured parameters. These may be thought of as single, double, and triple coincidences, although in practice the most common approach is to compute the first three moments of the multiplicity distribution (moments analysis)[1] or to work directly with the number of 0, 1, and 2 multiplets (multiplet approach).[2] Three unknown parameters such as sample mass, M, and α, can then be calculated from the three measured parameters.

The goal of neutron multiplicity analysis is to correctly assay in-plant materials without any prior knowledge of the sample matrix. The availability of a third measured parameter will make this possible for many materials, including impure plutonium oxide, oxidized metal, and some categories of scrap and waste. If the samples contain moisture or other neutron moderating materials, it may still be possible to obtain good assays by using the ratio of neutron counts in the various rings of detectors in the multiplicity counter to estimate and correct for the emitted neutron energy spectrum. Table I summarizes some of the potential applications for multiplicity counters.

TABLE I. Some potential applications of neutron multiplicity counters to nuclear materials present in DOE facilities.

1. Impure plutonium oxide with unknown α-particle source strengths like Am or unknown (α,n) reaction rates in materials like F.
2. Moist plutonium oxide, where the moisture increases the (α,n) reaction rate and decreases the average energy of the neutron spectrum.
3. Impure plutonium metal with an oxidized surface, Mg impurities, or other (α,n) emitters that are present in unknown quantity.
4. Pyrochemical scrap materials such as spent salts or salt scrub buttons from direct oxide reduction, molten salt extraction, or electrorefining processes.
5. Plutonium-bearing waste materials with some multiplication and enough (α,n) reactions to significantly bias the coincidence count rate.
6. Uranium metals with irregular geometry, where active neutron coincidence counting is inaccurate because of self-multiplication.

II. EXISTING PROTOTYPE COUNTERS

There are a surprising number of design options for multiplicity counters. Multiplicity measurements with existing thermal neutron counters have been reported for a three ring Active Well Coincidence Counter (AWCC)[3] and for a High Level Neutron Coincidence Counter (HLNC-II)[4] operated with the Euratom Time Correlation Analyzer.[2] Measurements with new counters have been reported for the Los Alamos dual mode thermal neutron multiplicity counter,[5] a fast neutron counter prototype,[6] and the Australian Nuclear Science and

TABLE II. Important parameters for existing or prototype neutron counters that have been used for multiplicity measurements. The range of plutonium mass and assay precision is determined from Fig. 1, assuming 6% ^{240}Pu, 1000-s counting times, and equal numbers of spontaneous fission and (α,n) neutrons (α = 1).

Neutron Multiplicity Counter	Detection Efficiency (%)	Neutron Die-away (μs)	Electronic Stability (%)	Range of Mass (g Pu)	Assay Precision (%)
Three-ring AWCC[3]	38	55	±0.3	15-15 000	2-6
HLNC-II[4]	18	43	±0.03	15-15 000	4-10
Dual-mode ctr. (High-eff. mode)[5]	43	24	±0.03	15-15 000	1-2
Fast Neutron Counter[6]	7	0.03	±5	80-15 000	1-6
Australian hybrid ^{3}He/Gd-loaded Scint.[7]	36	11.3	±3	15-1000	6-11

Technology Organization (ANSTO) prototype multiplicity counter.[7] Some of the important features of these prototypes are summarized in Table II. Other plastic- or liquid-scintillator-based options are also being evaluated.

Multiplicity measurements were first reported for the three-ring AWCC, which was used in the passive mode to assay a variety of plutonium samples.[3] Neutron detection efficiency is high for this counter, which contains 60 ^{3}He tubes, but the die-away time is also relatively high. Electronic stability is good because the thermal neutron capture process makes it easy to set a threshold below most of the expected detector pulse heights. The range of plutonium mass and the associated range of assay precision reported in Table II for this counter are based not on the actual measurements, which were analyzed with the multiplet approach, but rather on the results of the figure-of-merit code described in Sec. III below and illustrated in Fig. 1, which is based on the moments approach. The moments approach yields a significantly lower assay variance because more of the available data are used.

The HLNC-II contains 18 ^{3}He tubes and is undermoderated to reduce its weight.[4] Although this counter was designed for portable applications and was not intended for multiplicity measurements, it is included in this comparison because some multiplicity measurements have been carried out by Merlyn Krick at Los Alamos. If the multiplicity data are analyzed by the moments approach, it is possible to obtain an assay precision of 4 to 10% in 1000 s for some samples. Thus the HLNC-II could be used for the verification of some outliers by remeasuring them for several hours. Both the HLNC-II and the dual-mode counter described below are very stable because of their Amptek preamp-discriminator circuitry.

The dual-mode neutron multiplicity counter is the first counter designed specifically for multiplicity measurements.[5] It is designed to have both high neutron detection efficiency and low die-away time. This is achieved by using 130 ^{3}He tubes in five rings, with the tubes embedded in aluminum with only a thin sleeve of polyethylene. In the low-efficiency mode

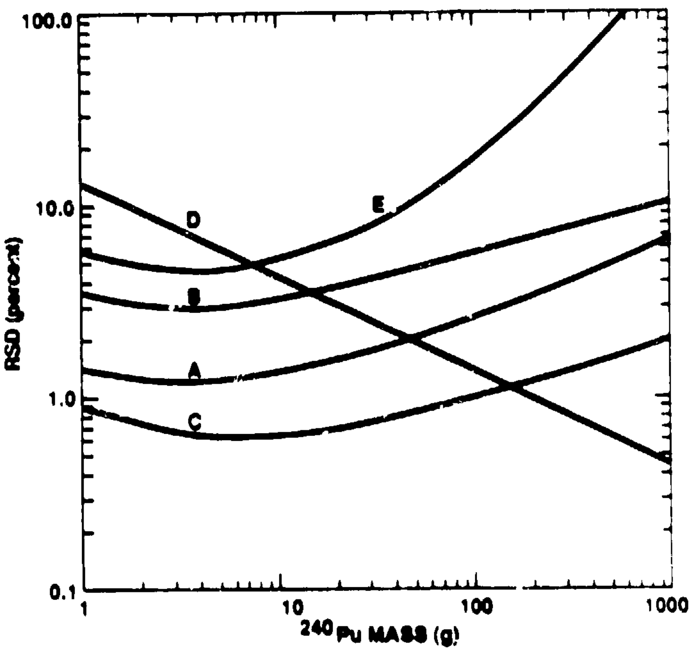

Fig. 1. Assay RSD in 1000 s as a function of ^{240}Pu mass for samples with α = 1. The curves (from Ref. 8) were calculated using an assay variance code, and are in good agreement with existing measurements analyzed with the moments approach. The curves are for the five prototype multiplicity counters described in the text: (A) three-ring AWCC, (B) HLNC-II, (C) dual-mode multiplicity counter, (D) fast neutron counter, and (E) ANSTO multiplicity counter.

of operation, each tube is wrapped with a cadmium insert, the neutron efficiency is 17%, and the die-away time is 11.8 μs. In the high-efficiency mode of operation (listed in Table II), the cadmium inserts are removed, the efficiency is 43%, and the die-away time is 24 μs. In this mode of operation, assay precisions of 1 to 2% can be obtained in 1000-s counting times for many samples, as illustrated in Fig. 1.

The fast neutron counter prototype[6] consists of four liquid scintillator cells, each 12.5 cm in diameter by 10 cm deep, and has a low efficiency because of the small solid angle of the cells. Pulse-shape discrimination eliminates fission gamma rays from the fast coincidence circuitry. The neutron die-away time is extremely low, about 20- to 30-ns flight time for the fast neutrons. Electronic stability is poor for scintillators because they measure a neutron recoil spectrum. In the future it should be possible to improve the stability of the fast neutron counter by using stabilization circuits or immersing the detectors in a large thermal mass. Assay precision for the fast neutron counter always improves with increasing sample mass: at low mass the precision is poor because of the low efficiency, but as the mass increases the precision always improves because of the very low die-away time.

Detection efficiency is also high for the Australian hybrid counter, which contains 80 liters of gadolinium-loaded liquid scintillator.[7] The gadolinium also acts as a neutron poison and thereby provides a low die-away time. Electronic stability is again not as good as for the thermal counters. Despite the high neutron detection efficiency and low die-away time, the assay precision is not good. However, this is caused by other effects that could be alleviated in the future: the ^{3}He trigger circuit has a very low efficiency, and the liquid scintillator has a high gamma-ray background rate.

III. ASSAY VARIANCE AS A FIGURE OF MERIT FOR MULTIPLICITY COUNTING

The importance of assay variance for multiplicity counters suggests its use as a figure of merit for comparing the performance of different designs. When other criteria such as cost, stability, and ruggedness have been met, assay variance may determine the ultimate usefulness of the instrument. This figure of merit can be used as an objective yardstick for comparing multiplicity counters and for predicting the effect of design changes.

A code has been developed to calculate assay variance from the factorial moments of the neutron multiplicity distribution.[8] The multiplicity distribution does not need to be measured, but can be predicted for any given sample from the design parameters of the counter. Comparison of the observed variance with the calculated variance shows that they are in good agreement for the available data. Then, for cases where data are not yet available, or for counters that are in the process of being designed, the figure-of-merit code can be used to predict the assay variance.

Figure 1 (from Ref. 8) shows assay relative standard deviation (RSD) as a function of ^{240}Pu mass for the current prototype counters.[3-7] The calculations assume 1000-s counting times and $\alpha = 1$ (equal number of spontaneous fission and (α,n) neutrons). Figure 1 shows that the counters with high neutron detection efficiency or low die-away time, or both, usually have the lowest assay variance for samples in the mass range of 1 to 1000 g ^{240}Pu. Efficiency and die-away time interact in a complex way to determine the assay variance. For example, Fig. 1 shows that the assay variance for a multiplicity counter first decreases, then increases with sample mass. For a conventional thermal neutron counter the assay variance always decreases and approaches an asymptotic value.

IV. DESIGN CRITERIA FOR NEUTRON MULTIPLICITY COUNTERS

From the measurements obtained to date with the above prototypes, and from the figure-of-merit code results in Fig. 1 and Ref. 8, we can identify some desirable design criteria for neutron multiplicity counters. These criteria are summarized in Table III, roughly in the order of their importance.

The multiplicity counter must have a high neutron detection efficiency to collect a sufficient number of high-order coincidences. Ideally, a useful neutron multiplicity counter should also retain the assay speed of current neutron coincidence counters. At the present time a practical goal is 1% RSD in 1000 s. The limiting factor in meeting this goal is the difficulty in obtaining 1% RSD on the triples, or third moment, and this same factor rules out the extraction of a fourth parameter from the multiplicity distribution.

The multiplicity counter should also have a short die-away time to optimize the ratio of the correlated signal to the uncorrelated background of accidental coincidences. As α increases, the need for short die-away time becomes more important. Figure 2 illustrates assay RSD at $\alpha = 10$ for the five prototype counters described in this paper. At this high value of α, such as might be found in some pyrochemical process residues, the assay RSD for all five counters becomes much higher, and the fast neutron counter (curve D) is now expected to have the lowest RSD for 10- to 1000-g ^{240}Pu samples. Thus, at high α, short die-away time becomes the most important criterion, and a fast neutron counter may be needed.

Three other design criteria involving neutron detection efficiency affect assay bias rather than assay precision. To the extent to which these criteria can also be met, the multiplicity counter will be more versatile and will require a smaller number of calibration curves for different material types. One criterion is the need for uniform detection efficiency across the sample volume. Secondly, the design should have a nearly optimal thickness of polyethylene so that small neutron energy spectrum shifts, such as those caused by sample moisture, will not bias the assay. Thirdly, the detection efficiency should in

TABLE III. Design criteria for neutron multiplicity counters, roughly in order of importance.

1	High neutron detection efficiency
2	Low neutron die-away time
3	Constant neutron detection efficiency over sample volume
4	Optimum detector moderator thickness
5	Efficiency independent of emitted neutron energy spectrum
6	Stable to at least 1%
7	Rugged and transportable
8	Simple operating and maintenance procedures
9	Moderate procurement cost

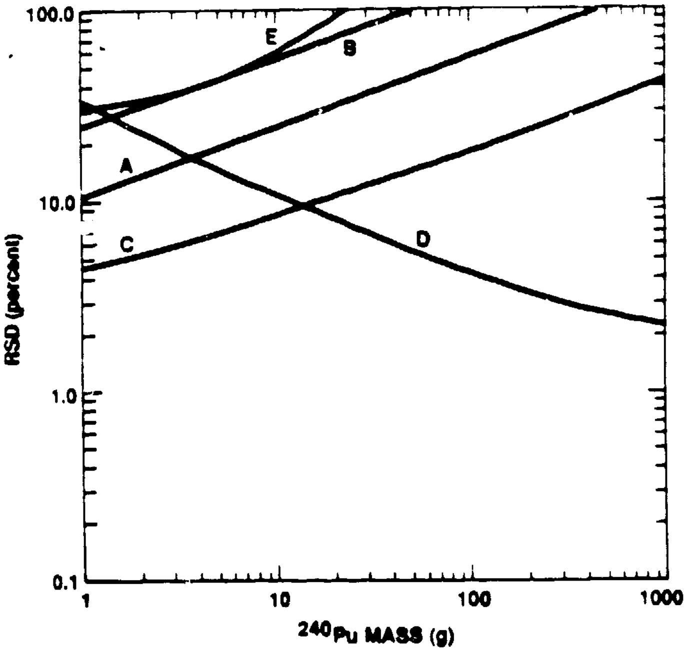

Fig. 2. Assay RSD in 1000 s as a function of ^{240}Pu mass for samples with $\alpha = 10$. The curves are for the five prototype multiplicity counters described in the text: (A) three-ring AWCC, (B) HLNC-II, (C) dual-mode multiplicity counter, (D) fast neutron counter, and (E) ANSTO multiplicity counter.

general be independent of energy spectrum shifts caused by (α,n) neutrons, which are emitted with energies in the range of 1 to 5 MeV. These three requirements are to some extent in conflict with the need for high efficiency and short die-away time, and thereby complicate the design process. However, if these criteria are not met, then neutron detection efficiency becomes a fourth unknown variable.

The neutron multiplicity counter electronics should be stable to 1% or better, a requirement easily met by existing thermal neutron counter electronics packages based on Amptek integrated preamp/discriminators. For scintillator-based counters this is a difficult criterion to meet, and will require the use of special stabilization circuitry.

For in-plant measurements by DOE or International Atomic Energy Agency inspectors, the multiplicity counter should also be rugged enough to be transportable from one process area to another, at least during the initial evaluation phase. This criterion is again easily met by thermal neutron counter designs, but is difficult to meet for liquid-scintillator-based fast neutron counters.

Operational simplicity, ease of maintenance, and moderate cost are other desirable criteria. Simplicity and maintainability are somewhat related to electronic stability and physical ruggedness, and the comments made above also apply here. Multiplicity counters will cost more than conventional counters because more neutron detectors, electronics, and software will be required. This will properly limit their use to measurement problems for which conventional counters are insufficiently accurate, such as those itemized in Table I. If multiplicity counters can truly provide assays that are independent of most matrix effects, the additional costs will be justified.

We are currently in the process of designing a new 3- or 4-ring thermal neutron multiplicity counter for in-plant measurement problems. The new design will be similar in concept to the 5-ring prototype developed by Krick, Bosler, and Swansen,[5] but will have a larger sample well and will try to meet as many of the above-mentioned design criteria as possible. We plan to begin an evaluation of this counter on in-plant materials such as impure oxides and pyrochemical residues in 1990.

V. CONCLUSIONS

From Fig. 1, we can conclude that several of the above prototypes give fairly good performance for some values of M and α, which is very encouraging. All can assay at least one or two samples overnight. The performance of two of the prototypes[6,7] can be improved significantly by easily attainable increases in the neutron detection efficiency or neutron trigger efficiency.

However, for most samples in the range of 1 to 100 g ^{240}Pu, the goal of 1% RSD in 1000 s can be met only by the short-die-away-time thermal neutron counter developed by Krick, Bosler, and Swansen[5] and operated with shift-register-based multiplicity electronics.[9] This design concept is thus the most likely candidate for the coming generation of in-plant multiplicity counters. This approach provides the best overall assay RSD at present and uses field-tested ^3He neutron detectors and associated electronics. The new thermal counter that we are now designing will be optimized for in-plant use and will be applied to as many materials as possible to push its usefulness to the limit.

For values of α larger than about 5, a scintillator-based fast neutron counter may have a lower assay RSD, as illustrated in Fig. 2. Research on plastic- or liquid-scintillator options is continuing at a slower pace because of problems with electronic stability and sensitivity to the neutron energy spectrum. Field-worthy fast neutron multiplicity counters are, perhaps, still one more generation removed.

For very high values of α (above 10), such as those found in americium- or fluorine-bearing salts or plutonium fluoride, multiplicity counters do not have a low enough assay RSD because of the high background signal. The sample self-interrogation technique, which can be accurate to 5 or 10% (Ref. 10) may be best for such materials. The choice of the best assay technique for a particular measurement problem depends on sample mass, multiplication, and α, with α being perhaps the dominant influence. Table IV lists the currently available neutron coincidence counting techniques and gives the range of α for which they are best suited. This table provides only rough guidelines, and more in-plant measurement experience is required to better define the range of applicability for each technique.

Until neutron multiplicity counters become available, we recommend that the neutron totals and neutron coincidence information currently provided by existing in-plant counters be fully exploited by applying existing known-α, known-M, or self-interrogation techniques. These techniques can provide better assay accuracy or new diagnostic information for many material categories.

TABLE IV. Presently available neutron coincidence counting techniques and their range of applicability as a function of the ratio of sample (α,n) neutron rate to spontaneous fission neutron rate, α.

Neutron Coincidence Counting Technique	α
Conventional coincidence counting with self-multiplication correction for samples with well-characterized composition (known α)	0-1
Conventional coincidence counting for samples with well-characterized geometry (known M)	0-5
Thermal neutron multiplicity counting	0-5
Fast neutron multiplicity counting	0-10
Sample self-interrogation technique	10-100

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This paper describes the work of many other people in addition to the author, although the conclusions are those of the author and are not necessarily shared by all other workers in the field. At Los Alamos, Merlyn Krick, Gene Bosler, and Jim Swansen have pioneered the development of thermal neutron multiplicity counters, and Joe Wachter has helped with the development of the fast neutron counter prototype. In Australia, John Boldeman and Nick Dytlewski have developed the gadolinium-loaded liquid scintillator multiplicity counter. In Europe, Walter Hage (Ispra), D. M. Cifarelli (Milan), and Klaus Boehnel (Karlsruhe) have developed the mathematical expressions needed to interpret the multiplicity distributions in moments form.

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